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Examiner: C. Seccuro, Jr.

Applicant(s): Winter et al.

Attorney Docket: HOE 92/F 294

For: POLYOLEFIN MOLDING COMPOSITION HAVING A BROAD MELTING RANGE, PROCESS FOR ITS PREPARATION, AND ITS USE

Assistant Commissioner for Patents
Washington, D.C. 20231

ATTENTION: Board of Patent Appeals and Interferences

APPELLANTS' BRIEF (37 C.F.R. 1.192)

Sir:

This brief, transmitted in triplicate, is in furtherance of the Notice of Appeal filed in this case on October 16, 1995. The fees required under 37 C.F.R. §1.17(f) and any required petition for extension of time for filing this brief therefor are dealt with in the accompanying "Transmittal of Appeal Brief".

This brief contains these items under the following headings and in the order set forth below:

- I. STATUS OF CLAIMS
- II. STATUS OF AMENDMENTS

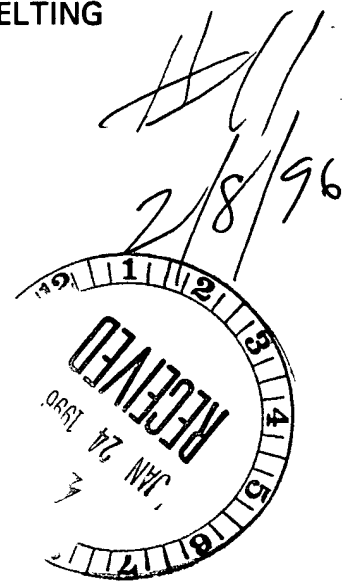
CERTIFICATE OF MAILING (37 CFR 1.8a)

I hereby certify that this correspondence (along with any paper referred to as being attached or enclosed) is being deposited with the United States Postal Service on the date shown below with sufficient postage as first class mail in an envelope addressed to the: Commissioner of Patents and Trademarks, Washington, D.C. 20231.

Date: January 15, 1996

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- III. SUMMARY OF INVENTION
- IV. ISSUES
- V. GROUPING OF CLAIMS
- VI. ARGUMENTS
 - A. Rejections Under 35 U.S.C. §102
 - B. Rejections Under 35 U.S.C. §103
- VII. APPENDIX OF CLAIMS INVOLVED IN THE APPEAL

The final page of this brief bears the attorney's signature.

I. STATUS CLAIMS

A. TOTAL NUMBER OF CLAIMS IN APPLICATION

Claims 1-3, 7, 8 and 11-16 are pending in this application.

B. STATUS OF ALL CLAIMS

Claims 1-3, 11, 13 and 14 are withdrawn from consideration but not canceled. In Appellants' Amendment, mailed February 1, 1995, claim 6 was canceled. Claims 7, 8, 12, 15 and 16 are rejected.

C. CLAIMS ON APPEAL

Claims 7, 8, 12 and 15. Appellants are not appealing the final rejection of claim 16.

II. STATUS OF AMENDMENTS

In Appellants' Amendment, mailed February 1, 1995, claim 6 was canceled. In the Final Rejection (Paper No. 9), mailed May 15, 1995, the Examiner correctly identified the pending claims as 1-3, 7, 8 and 11-16 but incorrectly included canceled claim 6 in the group of rejected claims 6-8, 12, 15 and 16. Rather, the rejected claims are 7, 8, 12, 15 and 16. A Notice of Appeal was mailed on October 16, 1995.

III. SUMMARY OF INVENTION

The present invention is directed to a process for preparing a polyolefin molding composition that is characterized by a narrow molecular weight distribution M_w/M_n and the following specific melt behavior:

- (i) a broad, bimodal or multimodal melting range in the DSC spectrum,
- (ii) where the melting range maximum is between 120 and 165°C,
- (iii) the half-intensity width of the melting peak is broader than 10°C and
- (iv) the width determined at quarter peak height is greater than 15° C.

The claimed process itself is characterized by the polymerization or copolymerization of at least two polyolefins by use of at least two different metallocenes. The melting points of the two polyolefin components must differ by at least 5°C.

The claimed invention is distinguishable over the prior art by Appellants' discovery that a mixture of at least two different metallocenes, each of which gives polyolefins having different melting points, gives a polyolefin product that does not, as expected, have a mixed melting point or a melting point below the melting point of the lower melting component. Advantageously, the polymer product of the claimed process has two melting points (Specification, page 1, line 27, to page 2, line 4).

IV. ISSUES

- (1) Whether claims 7, 8, 12 and 15 are unpatentable over 35 U.S.C. §102(b) over EP-A 0 310 734.
- (2) Whether claims 7, 8, 12 and 15 are unpatentable over 35 U.S.C. §103 over EP-A 0 310 734.
- (3) Whether claims 7, 8, 12 and 15 are unpatentable over 35 U.S.C. §103 over EP-A 0 128 046 and WO 90/03414, optionally further taken with EP-A 0351 189.

V. GROUPING OF CLAIMS

Claims 7, 8, 12 and 15 stand or fall together for each ground of rejection.

VI. ARGUMENTS

VI.A. Rejection Under 35 U.S.C. §102

Claims 7, 8, 12 and 15 are rejected under 35 U.S.C. §102(b) as allegedly anticipated by EP-A 0 310 734.

For support, the Examiner relies on Examples B, D and 7 of EP '734 (Table 1). In Example B, a Hf-containing catalyst was used in a polymerization process to produce a polymer having a melting point of 143°C. In Example D, a Zr-containing catalyst was used in a similar polymerization process to prepare a polymer having a melting point of 138°C, i.e, a melting point that is 5°C lower than the melting point of the polymer of Example B. The Examiner concludes, therefore, that it would be reasonable to expect that the polyolefin mixture of Example 7, which employs the Hf-catalyst of Example B and the Zr-catalyst of Example D, would consist of polymer components having melting points which differ by at least 5°C.

Firstly, Appellants submit that there is no inconsistency between the admission that a polyolefin having a certain melting point is formed due to the stereospecificity of each type of metallocene catalyst (Specification, page 12, lines 28-30), and the following statement of Appellants' invention:

Surprisingly, it has now been found that a mixture of at least two metallocenes each of which gives polyolefins of very different melting points give a polyolefin mixture which does not, as expected, have a mixed melting point or a melting point below the melting point of the lower-melting component, but instead has two melting points (Specification, page 1, line 30, to page 2, line 4).

EP '734 corroborates the prior art expectation that the melting point of a polymer product of a polymerization process employing two different metallocenes is below the melting point of the lower-melting component. Specifically, the polymer products of Examples 4-7 of EP '734 have a melting point below the melting point of the lower melting component.

Contrary to this expectation, Appellants have discovered that the polymer products of the claimed process do not have a mixed melting point or a melting point below the melting point of the lower melting component. Instead, it is Appellants' discovery that the claimed process produces polyolefin mixtures having two melting points, neither of which is below the melting point of the lower melting component. Additionally, the melting points of the polymer products of EP '734 were derived from DSC data but only one, the DSC peak temperature, is given in Table 1. There is no suggestion that the DSC measurements picked up a broad, bimodal or multimodal melting range.

Moreover, EP '734 neither expressly nor inherently teaches that the difference between melting points of the polymer components must be at least 5°C. This feature of the claimed process is necessary to the production of a polyolefin molding composition having the melt behavior set forth in claim 12. Polyolefin molding compositions having the desired melt behavior are highly suitable for molding applications, e.g., thermoforming, blow molding, etc., which applications are inappropriate for a polyolefin having a sharp melting and crystallization range (Specification, page 1).

The Examiner has correctly observed in Table 1 of EP '734 that the Hf- and Zr-catalysts of Examples B and D separately produce polymers having melting points which differ by 5°C. Appellants argue that it is equally as compelling to note that the difference in melting point of the product of Example A (134°C), which was prepared using a Hf-containing catalyst, and the melting points of the products of Examples C and D (137°C and 138°C), which were prepared using a Zr-containing catalyst, is less than 5°C. Furthermore, each of the polymer products of Examples 4-7 of EP '734 has a reported melting point below the melting point of the lower melting component.

Thus, each and every limitation of the claimed invention is not found within the four corners of EP '734. The reference is directed to a process for producing reactor blend polyolefins by using at least two different metallocene catalysts. However, EP '734 does not provide any meaningful teaching that the difference in melting points of the polymer components must be at least 5°C in order to obtain a

molding composition that exhibits a broad, bimodal or multimodal melting range in the DSC spectrum. This melting behavior is necessary for the production of polyolefins that are particularly suitable for thermoforming, blow molding, extrusion, injection stretch blow molding and certain film applications. An appreciation of the significance of the melt behavior set forth in claim 12 for polyolefin molding compositions is not embraced within the four corners of EP '734.

Accordingly, the rejection under 35 U.S.C. §102(b) is improper and withdrawal thereof is requested.

VI.B. Rejections Under 35 U.S.C. §103

- (1) Alternatively, claims 7, 8, 12 and 15 are rejected under 35 U.S.C. §103 as allegedly being obvious over EP '734.

Appellants repeat that EP '734 offers no suggestion that the difference between the respective melting points of the polymer components must be at least 5°C. Rather, Table I of EP '734 suggests that the difference between the melting points of the polymer components can be less than 5°C. The DSC data of EP '734 does not suggest that the polyolefin products of EP '734 are characterized by a broad, bimodal or multimodal melting range in the DSC spectrum. In fact, EP '734 teaches away from the claimed invention because, in Table 1, the polymer products of Examples 4-7 have a peak melting temperature which is lower than the peak melting point of the lower melting component.

Therefore, Appellants submit that claims 7, 8, 12 and 15 are not obvious over EP '734 and withdrawal of the rejection on the alleged basis is respectfully requested.

- (2) Claims 7, 8, 12 and 15 are rejected under 35 U.S.C. §103 as being unpatentable over EP 0 128 046 and WO 90/03414, optionally further taken with EP 0 351 189.

Both EP '046 and WO 90/03414 disclose a catalyst system of two or more different metallocenes for the production of polyolefins. However, EP '046 and WO 90/03414 are not concerned with the melt behavior of the polymer product. As such, there is no measurement of the melting point of the polymer product or the

polymer components. Accordingly, there is no teaching or suggestion that the melting points of the polymer components must differ by at least 5°C. As demonstrated by Table 1 of EP '734, without an express teaching, the difference in melting points of the polymer components could be less than 5°C. Furthermore, it is not suggested that the polymer products have a mixed melting point or a melting point that is not below the melting point of the lower melting component. Therefore, the references EP '046 and WO 90/03414 by themselves do not disclose or suggest the claimed process for the preparation of a polyolefin molding composition having the unique melt behavior set forth in claim 12.

Appellants respectfully submit that the Examiner's reliance on the publication Kaminsky, W., Schlobohm, M., Elastomers by Atactic Linkage of α -Olefins Using Soluble Ziegler Catalysts, Makromol. Chem., Macromol. Symp. 4, 103-118, pp. 103-104 (1986) is either misplaced or exceptionally limited when considered in view of the claimed process. The polymerization process described in the publication utilized a single Ziegler catalyst as opposed to the catalyst system of the claimed invention which comprises a mixture of at least two different metallocenes. Therefore, the significance of the publication can only be that the incorporation of hexene-1 into the polyethylene matrix LLDPE has, in general, an effect on melt behavior. However, this broad observation is of limited relevance to the distinctive features of the claimed process. Specifically, there is no teaching or suggestion that (1) the respective melting points of the polymer components differ by at least 5° and that (2) the polymer product possesses the melt behavior set forth in claim 12 and that (3) the polymer mixture has a mixed melting point or a melting point that is not below the lower melting component.

EP '189 discloses the preparation of an ethylene- α -olefin copolymer comprising ethylene and one or more α -olefin. The thermogram of the copolymer as determined by DSC shows an endothermic peak (a) in the range of from 75°-100°C and an endothermic peak (b) in the range of from 120°-140°C. The copolymer is polymerized in the presence of a catalyst system composed of a vanadium compound represented by the formula $\text{VO}(\text{OR})_n\text{X}_{3-n}$ (page 7, line 29). Thus, EP '189 provides no teaching or suggestion of a polymerization or copolymerization process which uses

a metallocene catalyst system. Accordingly, Appellants respectfully submit that there is no motivation to combine EP '189 with either EP '046 or WO 90/03414, which require a catalyst system comprising two or more different metallocenes to arrive at the claimed invention. The system for preparing the copolymer of EP '189 is so technically diverse from the metallocene catalyst systems of EP '046 and WO 90/03414 that, absent hindsight reconstruction of the claimed invention, there is no sound technical reason for the cited combination.

Thus, whether taken alone or in combination with EP '189, EP '046 and WO 90/03414 do not render the claimed invention obvious. Accordingly, withdrawal of the rejection of claims 7, 8, 12 and 15 under 35 U.S.C. §103 over EP '046 and WO 90/03414, optionally further taken with EP '189, is requested.

VII. APPENDIX OF CLAIMS

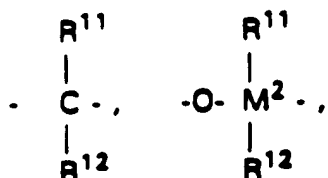
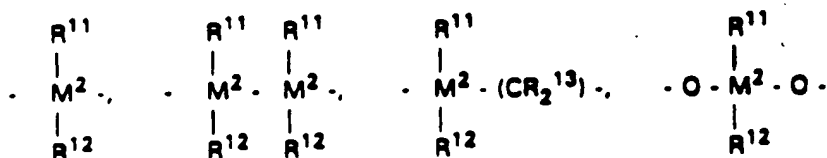
7. The process as claimed in claim 12, wherein

M^1 is Zr or Hf,

R^1 and R^2 are identical or different and are a hydrogen atom, a C_1 - C_3 -alkyl group, a C_1 - C_3 -alkoxy group, a C_6 - C_8 -aryl group, a C_6 - C_8 -aryloxy group, a C_2 - C_4 -alkenyl group, a C_7 - C_{10} -arylalkyl group, a C_7 - C_{12} -alkylaryl group, a C_8 - C_{12} -arylalkenyl group, or chlorine,

R^3 and R^4 are identical or different, monocyclic or polycyclic, unsubstituted or substituted hydrocarbon radicals which, together with the metal atom M^1 , can form a sandwich structure,

R^5 is



$=BR^{11}$, $=AIR^{11}$, $-Ge-$, $-Sn-$, $-O-$, $-S-$, $=SO-$, $=SO_2$, $=NR^{11}$, $=CO$, $=PR^{11}$ or $=P(O)R^{11}$, where R^{11} , R^{12} and R^{13} are identical or different and are a hydrogen atom, a halogen atom, a C_1 - C_4 -alkyl group, CF_3 group, a C_6 - C_8 -aryl group, a pentafluorophenyl group, a C_1 - C_4 -alkoxy group, a C_2 - C_4 -alkenyl group, a C_7 - C_{10} -arylalkyl group, a C_8 - C_{12} -arylalkenyl group or a C_7 - C_{12} -alkylaryl group, or R^{11} and R^{12} or R^{11} and R^{13} , in each case together with the atoms connecting them, form a ring,

M^2 is silicon or germanium,

R^8 and R^9 are identical or different and are as defined for R^{11} ,

m and n are identical or different and are zero or 1, where m plus n are zero or 1.

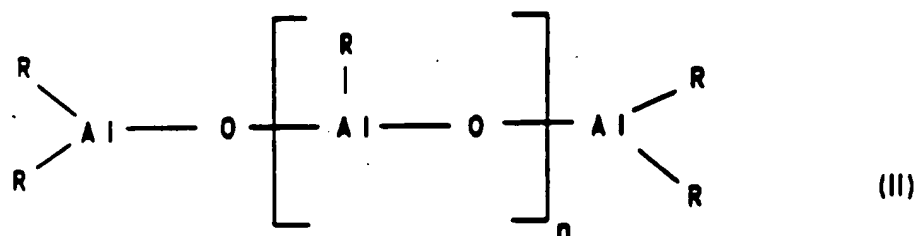
8. The process as claimed in claim 12, wherein
 M^1 is zirconium or hafnium,
 R^1 and R^2 are identical and are methyl or chlorine,
 R^4 and R^3 are indenyl, cyclopentadienyl or fluorenyl, where these ligands may carry additional substituents as defined for R^{11} , R^{12} and R^{13} , where the substituents may be different and, with the atoms connecting them, may also form rings,
 R^5 is a



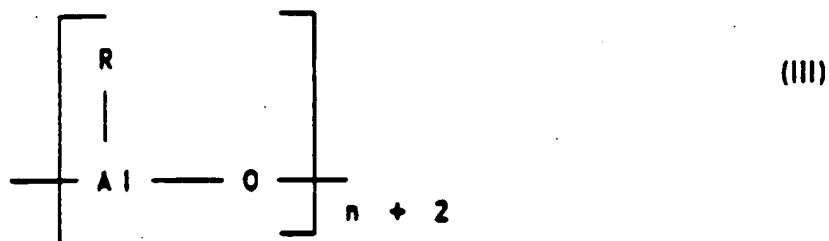
radical, and n plus m are zero or 1.

12. A process for the preparation of a polyolefin molding composition having a broad, bimodal or multimodal melting range in the DSC spectrum, where the melting range maximum is between 120 and 165°C, the half-intensity width of the melting peak is broader than 10°C and the width determined at quarter peak height is greater than 15°C, wherein such process comprises direct polymerization or copolymerization of at least two polyolefins of different melting point, where the melting points must differ by at least 5°C, and wherein the olefins have the formula $R^aCH=CHR^b$, in which R^a and R^b are identical or different and are a hydrogen atom or an alkyl radical having 1 to 14 carbon atoms, or R^a and R^b , together with the atoms connecting them, can form a ring, and are polymerized at a temperature of from -60 to 200°C, and a pressure of from 0.5 to 100 bar, in solution, in suspension or in the gas phase, in the presence of a catalyst, where the catalyst comprises at least

two metallocenes as transition-metal components and an aluminoxane of the formula II

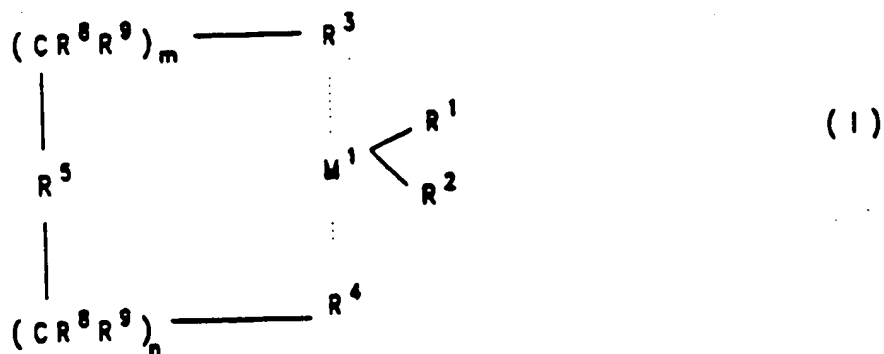


for the linear type and/or of the formula III



for the cyclic type, where, in the formulae II and III, the radicals R may be identical or different and are a C₁-C₆-alkyl group, a C₁-C₆-fluoroalkyl group, a C₆-C₁₈-aryl group, a C₆-C₁₈-fluoroaryl group or hydrogen, and n is an integer from 0 to 50, and the aluminoxane component may additionally contain a compound of the formula AlR₃,

where the transition-metal component used comprises at least two metallocenes of the formula I:



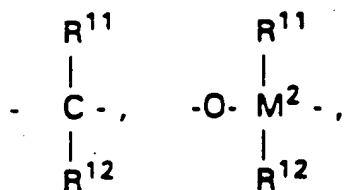
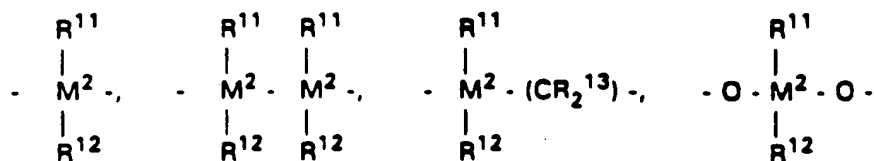
in which

M^1 is Zr, Hf or Ti,

R^1 and R^2 are identical or different and are a hydrogen atom, a C_1 - C_{10} -alkyl group, a C_1 - C_{10} -alkoxy group, a C_6 - C_{10} -aryl group, a C_6 - C_{10} -aryloxy group, a C_2 - C_{10} -alkenyl group, a C_7 - C_{40} -arylalkyl group, a C_7 - C_{40} -alkylaryl group, a C_8 - C_{40} -arylalkenyl group, or a halogen atom,

R^3 and R^4 are identical or different and are a monocyclic or polycyclic, unsubstituted or substituted hydrocarbon radical which, together with the metal atom M^1 , can form a sandwich structure,

R^5 is



$=Br^{11}$, $=AlR^{11}$, $-Ge-$, $-Sn-$, $-O-$, $-S-$, $=SO$, $=SO_2$, $=NR^{11}$, $=CO$, $=PR^{11}$ or $=P(O)R^{11}$,

where

R^{11} , R^{12} and R^{13} are identical or different and are a hydrogen atom, a halogen atom, a C_1 - C_{10} -alkyl group, a C_1 - C_{10} -fluoroalkyl group, a C_6 - C_{10} -aryl group, a C_6 - C_{10} -fluoraryl group, a C_1 - C_{10} -alkoxy group, a C_2 - C_{10} -alkenyl group, a C_7 - C_{40} -arylalkyl group, a C_8 - C_{40} -arylalkenyl group or a C_7 - C_{40} -alkylaryl group, or R^{11} and R^{12} or R^{11} and R^{13} , in each case together with the atoms connecting them, form a ring, and

M^2 is silicon, germanium or tin,

R^8 and R^9 are identical or different and are as defined for R^{11} ,
m and n are identical and are zero.

15. The process as claimed in claim 12, wherein the metallocenes are selected from the group consisting of $\text{rac-Me}_2\text{Si}(2\text{-methyl-1-indenyl})_2\text{ZrCl}_2$, $\text{rac-Me}_2\text{Si}(\text{indenyl})_2\text{HfCl}_2$, $\text{phenyl(methyl)Si}(2\text{-methyl-1-indenyl})_2\text{ZrCl}_2$, $\text{Me}_2\text{Si}(2\text{-methyl-4-phenyl-1-indenyl})_2\text{ZrCl}_2$, $\text{Me}_2\text{Si}(2\text{-methyl-1-indenyl})_2\text{ZrCl}_2$, $\text{Me}_2\text{Si}(\text{indenyl})_2\text{HfCl}_2$, $\text{phenyl(methyl)silyl}(\text{indenyl})_2\text{HfCl}_2$, $\text{rac-ethylene}(2\text{-methyl-1-indenyl})_2\text{ZrCl}_2$, $\text{rac-Me}_2\text{Si}(2\text{-methyl-4-phenyl-1-indenyl})_2\text{ZrCl}_2$, $\text{rac-ethylidene}(2\text{-methyl-4,6-diisopropyl-1-indenyl})_2\text{ZrCl}_2$, $\text{rac-Me}_2\text{Si}(2\text{-methyl-4,5-benzoidenyl})_2\text{ZrCl}_2$, $\text{dimethylmethylene}(9\text{-fluorenyl})(\text{cyclopentadienyl})\text{ZrCl}_2$, $\text{phenyl(methyl)methylene}(9\text{-fluorenyl})(\text{cyclopentadienyl})\text{ZrCl}_2$, $\text{rac-phenyl(methyl)silyl}(2\text{-methyl-4,6-diisopropyl-1-indenyl})_2\text{ZrCl}_2$, $\text{Ph(Me)Si}(2\text{-methyl-4-phenyl-1-indenyl})_2\text{ZrCl}_2$, $\text{rac-Me}_2\text{Si}(2\text{-methyl-4-(1-naphthyl)-1-indenyl})_2\text{ZrCl}_2$, $\text{rac-Me}_2\text{Si}(2,5,6\text{-trimethyl-1-indenyl})_2\text{ZrCl}_2$, $\text{rac-Me}_2\text{Si}(4,5\text{-benzo-1-indenyl})_2\text{ZrCl}_2$ and $\text{rac-Me}_2\text{Si}(4\text{-phenyl-1-indenyl})_2\text{ZrCl}_2$.

Respectfully submitted,



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